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**Title: Assessment and mitigation of the environmental burdens to air from land applied food-based digestate**

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### **Abstract (limit 150 words only)**

Anaerobic digestion (AD) of putrescible urban waste for energy recovery has seen rapid growth over recent years. In order to ascertain its systems scale sustainability, however, determination of the environmental fate of the large volume of digestate generated during the process is indispensable. This paper evaluates the environmental burdens to air associated with land applied food-based digestate in terms of primary pollutants (ammonia, nitrogen dioxide) and greenhouse gases (methane and nitrous oxide). The assessments have been made in two stages – first, the emissions from surface application of food-based digestate are quantified for the business as usual (BAU). In the next step, environmental burden minimisation potentials for the following three mitigation measures are estimated - mixed waste digestate (MWD), soil-incorporated digestate (SID), and post-methanated digestate (PMD). Overall, the mitigation scenarios demonstrated considerable  $\text{NH}_3$ ,  $\text{CH}_4$  and  $\text{N}_2\text{O}$  burden minimisation potentials, with positive implications for both climate change and urban pollution.

**Keywords:** *anaerobic digestion; bio fertilizer; digestate; environmental burdens; OFMSW*

**Capsule abstract:** In situ monitoring and analyses demonstrate the role of post-processing in greenhouse gases and air pollution mitigation from food-based digestate use as bio fertiliser.

### **Highlights:**

- In situ air pollution assessment of land applied digestate is performed.
- Environmental burden minimisation scenarios for digestate bio fertiliser presented.
- Food-based digestate show high ammonia volatilisation potential.
- Soil incorporated digestate effectively reduces  $\text{NH}_3$  but elevates  $\text{N}_2\text{O}$  emissions.
- Managing digestate emissions mitigate both climate change and air pollution.

## 1. Introduction

Digestate, the semi-solid residue obtained post biogas extraction in anaerobic digestion (AD), is considered a vital source of organic matter and nutrients, especially nitrogen. It is increasingly being applied as soil conditioner/amendment (on urban gardens, farmlands, recreation/sports grounds, fish ponds, etc.), or alternatively for developing energy crops on brownfield/marginal land and sports turf production (WRAP, 2013). This is mainly due to its two attributes - one, for providing a low carbon substitute for fossil fertilisers (Chambers and Taylor, 2013; WRAP, 2011); two, for restoring soil organic matter and for closed-loop nutrient recycling, especially mineral nitrogen (Fricke et al., 2007; Karagiannidis and Perkoulidis, 2009; Möller and Stinner, 2009). Consequently, digestate field application has been identified as a sustainable practice in terms of meeting the EU standards for good agricultural and environmental condition (GAEC) (RPA/Defra, 2012). However, with greater emphasis on strategies for diverting biowastes from landfill and their sustainable re-utilisation through valorisation in AD, the volumes of digestate are expected to increase rapidly (typical digestate represents 70-95% of the feedstock volume) (Lukehurst et al., 2010). Digested slurries have been found to be significant sources of ammonia ( $\text{NH}_3$ ), methane ( $\text{CH}_4$ ) and nitrous oxide ( $\text{N}_2\text{O}$ ) emissions (Amon et al., 2006; Bacenetti et al., 2013; Nkoa, 2014; Wulf et al., 2002a), with potential implications for local-to-regional climate (NRC, 2002; Ravishankara et al., 2009) and human health (Peel et al., 2013). Nkoa's (2014) detailed review has suggested that digestates can be considered as organic amendments (or organic fertilizers) only when properly handled and managed.

The content and quality of digestate depends largely on both the feedstock and the hydraulic retention time (HRT) of the digester; usually longer HRT reduces the organic content owing to more effective methanogenesis (Szűcs et al., 2006). Digestate quality is further affected by maturing in storage tanks (Menardo et al., 2011). Rigid compliance criteria for Class I digestate have been set by the European Commission (EC) and the British Standard Institution (BSI) (BSI, 2010; EC, 2014). Although the scale of AD operation dedicated to organic waste treatment is at an all-time high (and on the rise), there is still relatively little published information on the composition and potential environmental behaviour of digestate, particularly from AD plants processing food wastes (Tiwarý et al., 2015; Whelan et al., 2010). Anaerobic digestion of slurry tends to increase ammoniacal-N concentrations, reduce carbon to nitrogen ratios and increase pH, thereby increasing the risk of  $\text{NH}_3$  losses during storage and soil application (Möller and Stinner, 2009). Inappropriate disposal of unstable digestate on land may lead to formation of residual biogas, with potential health issues from exposure to its constituent non-methanic volatile organic compounds and other hazardous air pollutants and odorous compounds (Palmiotto et al., 2014).

During AD, the majority of organic (slow release) nitrogen is transformed into readily available nitrogen (RAN) (Defra, 2011), specifically for protein-rich feedstocks, including the organic fraction of the municipal solid waste (OFMSW)<sup>1</sup>, dairy by-products and slaughterhouse waste (Kryvoruchko et al., 2009; Menardo et al., 2011). Typical total-N content of food-based digestate ranges between 5-8 kg m<sup>-3</sup>, with about 60-80% of this present as RAN; the proportion of RAN to total-N in food-based digestate is nearly 40% higher than manure-based digestate (202 kg and 145 kg respectively for every 250 kg total-N) (WRAP, 2011). Moreover, for digestate from kitchen waste feedstock RAN of as much as 99% of corresponding Total-N has been reported (Furukawa and Hasegawa, 2006). Sensitivity analyses conducted to assess the split share of digestate total-N (i.e. organic-N, NH<sub>4</sub>-N and NO<sub>3</sub>-N) on acidification potential have reported NH<sub>3</sub> and NO<sub>3</sub> as the main contributors to the enrichment of pollutants in direct air and water environments respectively (Bernstad and la Cour Jansen, 2011; Evangelisti et al., 2014). Free ammonia concentration is affected mainly by temperature, pH and total ammoniacal nitrogen (TAN) concentration (Chambers and Taylor, 2013). Whereas a number of multivariate data analysis protocols using advanced sensor technologies have become available, the majority of these are currently (2014) limited to AD process monitoring; there are limited approaches for digestate quality monitoring and they are all based on offline instrumentation (Oppong et al., 2012). Minimum quality requirements for whole digestate, separated liquor and separated fibre have been prescribed as part of a Publicly Available Specification (PAS 110)<sup>2</sup> standards in the UK (BSI, 2010). PAS 110 Clause 10 provides guidance on obtaining representative samples of all three types of digestates (whole, fibre, liquor) via one or more sampling access points appropriately located in the digestate production/storage system prior to its use.

The knowledgebase involving environmental impacts from digestate land application is largely developed from traditional approaches for cattle manure management, which requires incorporating the unique and emerging characteristics of OFMSW digestate. From an environmental point of view there is an existing challenge of reducing the gaseous N-losses (NH<sub>3</sub>, NO<sub>2</sub>, N<sub>2</sub>O) from digestate soil application to the fields (Balsari et al., 2007; Misselbrook et al., 2005a; Nyord et al., 2008; Tiwary et al., 2015). This study aims to determine and interpret the seasonal emissions profile to air from digestate slurry applied to urban soil. The latter has been assessed through small plot experiments, applying digestate acquired from community-scale AD operations. It evaluates the implications for climate change and urban pollution in terms of fitness for purpose of the available practice, including some proposed mitigation strategies. Preliminary outputs from field trials of in situ monitoring of soil-applied digestate for air emissions of N-pollutants (NH<sub>3</sub>, NO<sub>2</sub>) and

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<sup>1</sup> OFMSW is defined by the European Commission as “biodegradable park and garden waste, food and kitchen waste from household, restaurants, caterers and retail premises and comparable waste from food processing plants”.

<sup>2</sup> PAS (Publicly Available Specification) is a sponsored fast-track standard driven by the needs of the client organisations in the United Kingdom, developed according to guidelines set out by British Standard Institute.

greenhouse gases (GHGs: CH<sub>4</sub>, N<sub>2</sub>O) are reported. These results feed into a review of approaches to mitigate the environmental burdens while promoting re-utilisation potentials of anaerobically digested slurry as marketable bio fertilisers.

## **2. Materials and methods**

### *2.1. Review of sampling and analytical techniques*

A review of available techniques was deemed essential for designing a robust assessment methodology for evaluating the emissions from both the baseline and the responses to plausible mitigation strategies.

#### *2.1.1 Primary air pollutants*

Several studies have reported monitoring of NH<sub>3</sub> emissions from land-applied manure in Europe (Gericke et al., 2011; Loubet et al., 1999; Misselbrook et al., 2005a; Nyord et al., 2012; Nyord et al., 2008), Australia (Leuning et al., 1985), USA (Parker et al., 2013) and Canada (King et al., 2012). Inter comparison of different monitoring techniques are also reported (NRC, 2002; Parker et al., 2013) (Misselbrook et al., 2005b). The following three broad categories of approaches have been applied for estimating air pollutant emission rates from area sources: i) Micrometeorological mass balance techniques, where fluxes are calculated from a large footprint using tower-based instrumentation (e.g., gradient methods, eddy covariance); ii) Indirect methods, where ambient concentrations are measured and source emission rates are either back-calculated using dispersion models or using known transfer factors from standards (also known as ‘standard comparison’ method; iii) Direct methods, where fluxes are measured from samples collected from the source using portable wind tunnels or flux chambers, followed by colorimetric/FTIR spectroscopy. Among these, micrometeorological mass balance methods have been found to provide more realistic estimates, thus widely used in field monitoring (Generemont et al., 1998; Nyord et al., 2012; Wilson and Shum, 1992), but limited to applications over larger source areas with sufficient fetch for integration of emission rates. Standard comparison methods are popular owing to their cost-effectiveness (Möller and Stinner, 2009; Wulf et al., 2002b) but are quite sensitive to surface characteristics and meteorological conditions, requiring regular corrections to the fluctuations in the transfer factor applied in emissions estimates. In particular, such methods assume the transfer factors estimated from standard plots to be valid for calculating the emission rates from the experimental plots, overlooking the inconsistencies in micrometeorological conditions, especially the wind fields between the standard and the experimental plots. Despite their limitations, portable wind tunnels and flux chambers are often the only available, direct method for assessing air pollutant fluxes from small individual area sources. Theoretical principles of wind tunnel technique have been described in detail in the literature (Loubet et al., 1999; Misselbrook et al.,

2005b). The appropriateness and accuracy of portable wind tunnels and flux chambers for quantifying area source emissions have been extensively discussed in the literature (Fowler et al., 2001; King et al., 2012; Loubet et al., 1999; Misselbrook et al., 2005b; Nyord et al., 2012; Parker et al., 2013).

Qualitative assessment of the cost-benefits of the different monitoring techniques have also been reported (see for example (Gericke et al., 2011; Misselbrook et al., 2005b)). For small-plot comparative measurements, a wind tunnel system is recommended, albeit with a caveat of its tendency to modify the temperature at the emitting surface relative to the ambient conditions owing to canopy effect (Braschkat et al., 1993; Misselbrook et al., 2005b; Nyord et al., 2012), thereby influencing the emission rates. Application of wind tunnel technique is thus far best suited for making comparative measurements for treatments imposed on small plots and less representative for estimating the absolute air emissions from large area sources.

#### 2.1.2. Greenhouse gases

Compared to monitoring of primary air pollutants, there appears to be a greater consistency in the reported literature on monitoring of greenhouse gases (GHG) from area sources. The majority of the studies have applied closed chambers (static or mobile) for sampling of N<sub>2</sub>O and CH<sub>4</sub>, followed by concentration estimations using gas chromatography (Amon et al., 2006; Wulf et al., 2002a). With closed chambers, however, reportedly there is an issue with negative feedback of increasing gas concentration in the chamber on the rate of diffusion of the gases, implying that the rate of release is often very high immediately after land application.

### 2.2. *Sampling and analysis*

#### 2.2.1 Experimental design

A dedicated wind tunnel experimental set up was developed following Lockyer (1984) and Loubet et al. (1999), comprising of an upwind-downwind gas sampling system and an air extractor (fitted with an anemometer to monitor the average wind speeds through the tunnel canopy). The front portion of the wind tunnel was fabricated using transparent polycarbonate material covering 1 m<sup>2</sup> (2 m long x 0.5 m wide), offering exposure to direct sunlight and ensuring negligible temperature interference from the tunnel to the treated surface area. Three separate wind tunnels were operated simultaneously in parallel (**Fig. 1**) to acquire statistically representative samples (Misselbrook et al., 2005a; Nyord et al., 2012). Air was drawn from the upwind/downwind sampling ports located on top of the tunnel into impinger tubes containing the absorbing media through three sets of polytetrafluoroethylene (PTFE) tubing (approximately 4 mm internal

diameter) at a flow rate of  $3 \text{ l min}^{-1}$  using an arrangement of two manifolds and suction pumps. For GHG, only the downwind port of the middle tunnel was used to draw the sample from an air-tight chamber inserted into the soil to a depth of 10 cm.

### 2.2.2 Field sampling

All land application experiments were conducted at a field site representative of tropical climates, located at The Energy and Resources Institute (TERI) campus (suburbs of New Delhi,  $28.45^\circ\text{N}$ ,  $77.03^\circ\text{E}$ ), India. Three wind tunnels, each covering an exposed area of  $1 \text{ m}^2$ , were operated in parallel on a  $4\text{ m} \times 4\text{ m}$  plot (**Fig. 1**), and predominantly composed of sandy loam soil ( $\text{pH} = 5.5$ , organic matter content (%)  $= 3.7$ ). The wind tunnels were positioned such that the canopy inlets were located at the upwind edge of the plots and they remained in the same position for the entire duration of each experiment. To avoid cross-interference of emissions between the neighbouring wind tunnels care was taken to restrict the spread of fresh digestate slurry within the exposed portion of each tunnel using a watering can. The amount of digestate spread on different plots was volume-limited to  $0.02 \text{ m}^3$  (i.e. 20 L), with a spread area of  $1 \text{ m}^2$  and application thickness of 2 cm per plot. All the experiments were conducted with no precipitation recorded within the first 24 h following digestate application.

Air pollutants ( $\text{NH}_3$ ,  $\text{NO}_2$ ) were sampled over 6 consecutive days following digestate application based on the literature (Nyord et al., 2012; WRAP, 2011). Although previous experiments using digested cattle slurry have reported  $\text{NH}_3$  emissions to cease after 48 h (Amon et al., 2006), a relatively longer sampling period was chosen for food-based digestate, potent to exhibit longer emission patterns owing to higher RAN (Chambers and Taylor, 2013; Whelan et al., 2010). A quality assurance run was performed to establish the sampling protocols for  $\text{NH}_3$  using two-serially connected impinger tubes to the downwind port. This experiment showed that less than 2% of the total  $\text{NH}_3$  trapped during the sampling reached the second impinger tube, which confirmed the emissions were insufficient to fully saturate the absorbing media in the first impinger tube. Therefore, for all subsequent experiments only one impinger tube was used for the entire sampling. During each experiment, the impinger tubes filled with absorbing media, and connected to the upwind/downwind sampling ports of all the three wind tunnels, were simultaneously exposed continuously at a stretch for 4 h (corresponding to sampling medians of 2, 26, 50, 74, 98 and 122 h).

Monitoring of greenhouse gases ( $\text{CH}_4$ ,  $\text{N}_2\text{O}$ ) was performed from an air-tight chamber connected to the downwind sampling port of the middle wind tunnel owing to logistical limitation in simultaneous operation of gas chromatograph. Following land application,  $\text{CH}_4$  was monitored for up to 7 days on a daily basis (Wulf et al., 2002a);  $\text{N}_2\text{O}$  was monitored for up to 30 days at a sampling interval of 0, 1, 2, 3, 5, 9, 16, 23



and 30 days, with reduced sampling frequency beyond the first week as per the literature (Amon et al., 2006; Wulf et al., 2002a).

### 2.2.3 Analysis

Methodological details of the analysis adopted for different digestate parameters (dry matter (DM) content, pH, total Kjeldahl N (TKN) content, total ammoniacal N (TAN) content, etc.) are provided in **Table 1**. Analyses of primary air pollutants followed the standard practice for quantifying ambient air concentrations by bubbling a known volume of air through the impinger tubes filled with absorbing media and connected to the upwind/downwind sampling ports of the tunnels.  $\text{NH}_3$  was quantified through the Indophenol method (Method 401, Air Sampling and Analysis, 3<sup>rd</sup> Edition, (CPCB, 2011)) using a dilute solution of sulphuric acid as absorbent to precipitate the airborne ammonia as ammonium sulphate followed by its colorimetric analysis by reaction with phenol and alkaline sodium hypochlorite to produce indophenol.  $\text{NO}_2$  was quantified through the modified Jacobs & Hochheiser method (IS5182 Part 6, Methods for Measurement of Air pollution: Oxides of nitrogen (CPCB, 2011)) by bubbling the sample through a solution of sodium hydroxide and sodium arsenite to convert the airborne  $\text{NO}_2$  into nitrite ion ( $\text{NO}_2^-$ ). In the subsequent step, this was colorimetrically analysed by reacting with phosphoric acid, sulphanilamide and N-(1-naphthyl)-ethylenediamine di-hydrochloride (NEDA). The emission per sampling period was calculated as the product of the volume of air passing through the tunnel and the difference in outlet and inlet air concentrations. For each measuring period, the background pollutant concentration from the inlet located on the upwind face of the tunnel was estimated and subtracted from the cumulative concentration estimated from the port located on the downwind face of the exposed area.

The GHGs were analysed by means of a gas chromatograph (GC-5700 series, Nucon Engineers, New Delhi) using a 1.8 m (~6 ft) long Poropak-Q column with thermal conductivity detector (TCD). The injector port, the detector and the oven were operated at temperatures of 40, 40 and 35°C respectively. Argon was used as a carrier gas at a pressure of 1.8 kg  $\text{cm}^{-2}$ . One ml of gas sample was injected using a micro syringe into the gas chromatograph for analysis.

### 2.3 Scenario analysis

The following scenarios were developed and tested sequentially using the experimental facility. Details of the physico-chemical properties of the digestate used in this analysis are presented in **Table 2**. For primary air pollutant monitoring ( $\text{NH}_3$ ,  $\text{NO}_2$ ), the cumulative emissions for the different scenarios were estimated as arithmetic means of three replicates (**Fig. 1**), as it is considered more robust (and less biased) for small

numbers of replicates (Wulf et al., 2002a). The overall emission trends were obtained by fitting regression curves to the experimental data in SigmaPlot v12.5 (Systat Software Inc.).

#### 2.3.1 Business as usual

As a first step, assessments were conducted for the baseline emission patterns (BAU) following land application of digestate obtained from processing 100% OFMSW in a two-stage AD plant, available at the study site. The digestate was relatively fresh when land applied, keeping the approach consistent with the growing emphasis on reducing the storage time of digestate used as soil amendments, in order to avoid nutrient (mainly RAN) and carbon loss.

#### 2.3.2 Mixed-waste digestate

Typically, mixed-waste feedstock (with higher C/N ratio) have tendency to improve biogas yield and lower NH<sub>3</sub> release potentials from digestate. Co-digestion of OFMSW with cattle manure is commonly adopted in wet digestion (Banks et al., 2011). On the other hand, co-digestion of waste having high N-content (food waste, vegetable waste, food processing industry waste and slaughterhouse waste) with waste paper (typical mix of 95% to 5% respectively) is proposed for controlled dry digestion (Li et al., 2011; Takata et al., 2013), owing to its collateral benefits of adjusting the C/N ratio of the medium and regulating the accumulation of both NH<sub>3</sub> and VFA in the reactor. As the first mitigation measure, performance of a mixed waste digestate (MWD), obtained following co-digestion of 70% OFMSW (food, fruit and vegetable waste) with 30% cattle dung, was evaluated to optimise the earmarked emissions.

#### 2.3.3 Soil incorporated digestate

Uniform application of digestate slurry near or under the soil surface is recommended as an environmental best practice in the literature (Amon et al., 2006; Nyord et al., 2012). However, ‘sub-surface incorporation’, has demonstrated mixed results in abatement of gaseous emissions. While it positively mitigates NH<sub>3</sub> emissions (Chambers and Taylor, 2013; Nyord et al., 2012; Wulf et al., 2002b), it tends to exacerbate the GHG emissions under anaerobic conditions (CH<sub>4</sub>, N<sub>2</sub>O) (Möller and Stinner, 2009; Nkoa, 2014; Wulf et al., 2002a). The proposed mitigation measure of soil incorporated digestate (SID) involved shallow injection (5-7 cm below ground) of the BAU digestate, followed by its immediate soil incorporation based on the best available technique (BAT) recommended in the literature (Brizio and Genon, 2010).

#### 2.3.4 Post-methanated digestate

Typically during the first 2 months of digestate storage post-methanation, up to 15% additional CH<sub>4</sub> yields have been reported (Balsari et al., 2013; Menardo et al., 2011; Weiland, 2010). However, digestate storage

during post-methanation also have collateral influence on  $\text{NH}_3$  emissions, owing to high ammonium nitrogen ( $\text{NH}_4\text{-N}$ ) concentration (Whelan et al., 2010). Previously reported  $\text{NH}_3$  emission rates ranged between 2.06 and 4.44 g  $\text{NH}_3 \text{ m}^{-2}$  and between 7.89 and 14.6 g  $\text{NH}_3 \text{ m}^{-2}$  from stored, whole digestate and from digested liquid fraction respectively (Gioelli et al., 2009). The post-methanated digestate (PMD) for this study was obtained after maturing the BAU digestate into a 150 L concrete tank with a floating dome for 45 days.

### 3 Results and discussion

The sampling and analyses steps described above were repeated to obtain the corresponding emissions for  $\text{NH}_3$ ,  $\text{NO}_2$ ,  $\text{CH}_4$  and  $\text{N}_2\text{O}$  in order to evaluate the environmental performance of the proposed mitigation scenarios MWD, SID and PMD with reference to the BAU. The following sections describe the observed trends as well as their seasonal variations, if any; these are discussed in the context of developing effective digestate handling and management strategies to achieve reduced environmental burdens and enhanced economic values, the latter in terms of improved nutrients and organic matter reutilisation.

#### 3.1 Emissions trends

##### 3.1.1 Ammonia emissions

The  $\text{NH}_3$  emissions for all the scenarios showed a common trend of highest emissions within the first two days and negligible emissions beyond the four-day threshold (**Fig. 2a**), which agrees with previous studies (Amon et al., 2006; Möller and Stinner, 2009; Wulf et al., 2002a). Preliminary results suggest that digestate surface application leads to significant losses of  $\text{NH}_3$  to air over a short span during the first week, with the business as usual having the highest cumulative  $\text{NH}_3$  emissions following land application, estimated to be over 65% of the applied TAN (**Fig. 2b**). Compared to this, application of mixed feedstock digestate and post-methanated digestate had cumulative emissions around 45% and 35% of applied TAN respectively, resulting in over 35%  $\text{NH}_3$  reductions over the business as usual from these interventions. However, the maximum  $\text{NH}_3$  abatement of around 85% was achieved from slurry soil incorporation immediately following application; the cumulative emissions estimated in this case was around 10% of the applied TAN and negligible  $\text{NH}_3$  emissions were observed from the second day onward ( $< 1\%$  of TAN), which was along the lines of previous reportings (Nyord et al., 2012; Wulf et al., 2002a).

### 3.1.2 Nitrogen dioxide emissions

The majority of AD literature has extensively reported on  $\text{NH}_3$  emissions whereas  $\text{NO}_2$  emissions from digestate application are not adequately accounted for in the emissions inventories.  $\text{NO}_2$  emissions from soil amended plots are considered highly uncertain, and strongly influenced by the soil microenvironment (MNP, 2007); estimates from a previous digestate assessment study suggest it to be up to 15% of the  $\text{NH}_3$  emissions in the Netherlands (de Vries et al., 2003). Results from our study showed overall trends for  $\text{NO}_2$  emissions similar to  $\text{NH}_3$ , with BAU having the largest cumulative emissions, followed by MWD while SID showed negligible  $\text{NO}_2$  emissions (**Fig. 3**). The rate of emissions for both BAU and MWD were very feeble past 24 h from land treatment, and their respective cumulative emissions were estimated within 10% and 4% of TAN.

### 3.1.3 Methane emissions

Compared to  $\text{NH}_3$  the observed  $\text{CH}_4$  emission intensity was noted to be relatively short-lived for BAU, with a steep decline in emissions from day 3 onwards (**Fig. 4**). Nevertheless, based on our analysis, in the initial 1-2 days from land application, BAU had the highest  $\text{CH}_4$  emissions intensity, followed by MWD. PMD, despite showing similar emissions trend temporally, had a much reduced intensity (up to 56% lower) compared to the BAU. This is mainly attributed to residual  $\text{CH}_4$  extraction from post-methantion; reported estimates of residual  $\text{CH}_4$  potentials during digestate maturing/long term storage (up to 180 day) vary, ranging from 5-15% to 12-31% of total methane production (Weiland, 2003). These variations are linked to the feed quality, the organic loading rate (OLR) and the HRT of the AD process, as well as the moisture content of the digestate itself, typically reported residual  $\text{CH}_4$  potential for animal manure, energy crops and food industry waste range between 2.88 and 37.63 L  $\text{kg}^{-1}$  volatile solids (Menardo et al., 2011). The steep hike in emissions in the initial phase (within 1-2 days) post-application for BAU, MWD and PMD is mainly attributed to the readily available dissolved  $\text{CH}_4$ , produced during storage of the substrate. On contrary, SID showed a lower emission intensity in the first 24 hour, peaking only after 2 days of land application. These trends of delayed peaking of  $\text{CH}_4$  from injected slurry is also found in previous monitoring campaigns (Wulf et al., 2002a) and can be mainly attributed to kick-starting of sub-surface anaerobic degradation of VFAs through methanogenesis under humid conditions.

### 3.1.4 Nitrous oxide emissions

Nitrous oxide is formed as an intermediate product of both nitrification and de-nitrification. Previous studies have reported strong increase in  $\text{N}_2\text{O}$  emissions from mitigation strategies for reducing  $\text{NH}_3$  volatilisation involving either application of liquid digestate, typically with a narrow C/N ratio and high soil infiltration levels (Möller and Stinner, 2009), or injection and soil incorporation of digestate (Wulf et al., 2002a).

Indirect N<sub>2</sub>O production from emitted NH<sub>3</sub> has been considered a potential indirect contributor to global warming (Wulf et al., 2002a), with approximately 1% of NH<sub>3</sub>-N assumed to be re-emitted to the atmosphere as N<sub>2</sub>O-N (IPCC, 2001). However, this mechanism was negated as a possible source for N<sub>2</sub>O in our experiments and all N<sub>2</sub>O was attributed to direct emissions, given the use of wind tunnel and the timescale of samples collected. The BAU and MWD showed similar levels of emissions, PMD had relatively lower emissions but SID showed a peculiar, and quite contrasting emission trend, with increasing levels of emissions recorded about two weeks post land application (**Fig. 5**), which is also reported in previous studies (Möller and Stinner, 2009; Nyord et al., 2008). The background N<sub>2</sub>O emissions from the bare soil have also been shown in this figure for reference.

### 3.2. Seasonal effects

There is evidence of increased air emissions following digestate soil application on environmental factors, such as ambient temperature, wind speed and precipitation (Chambers and Taylor, 2013; Nyord et al., 2012; Parker et al., 2013; Peel et al., 2013). This section reports outcomes of the air emissions evaluated for two contrasting periods: summer (August – October) and winter (January – March) (**Table 3**). Based on the outputs from the monitoring experiments, potential environmental burdens for NH<sub>3</sub>, NO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O to air were estimated over the two seasons as a function of the fresh matter digestate mass applied to soil for the BAU, MWD and SID scenarios (**Table 4**). Distinct seasonal characteristics influencing the emissions have been identified below.

**Sunlight hours** - Our results showed strong dependence of the monitored emissions on sunlight hours; the observed trends as NH<sub>3</sub> >> N<sub>2</sub>O > CH<sub>4</sub>, whereas NO<sub>2</sub> showed an inverse dependence. For all the trace gases the cumulative emissions remained unaltered and the effect of cooler and moist periods during winter mainly impeded their release rates, as reported in previous studies (Wulf et al., 2002a). NO<sub>2</sub> emissions were marked with large fluctuations and were found to be higher during overcast winter months (typically with average ambient temperature around 10°C and less than 2 hours of direct sunlight on the field plots).

**Wind speed** - In previous studies, wind speed has been reported as one of the parameters with the greatest influence on NH<sub>3</sub> emissions from slurries (Misselbrook et al., 2005a). As followed in this study, the wind tunnel set up reportedly provides a more robust wind-sensitive NH<sub>3</sub> emissions estimates (Rong et al., 2009). The contrasting wind micro-environment recorded over the two seasons, with up to 75% reduction in winter over summer (**Table 3**), seems to have resulted in large differences between the observed emissions, especially for NH<sub>3</sub>.

Soil moisture - Lower wind speed during winter resulted in sustained build-up of humid conditions inside the tunnels, enhancing the soil moisture content (**Table 3**). This in turn resulted in poor soil respiration, contributing to reduced emissions during this period. On contrary,  $\text{NH}_3$  losses were intensified during summer when slurries were applied to dry soils under warm weather conditions. For  $\text{CH}_4$ , the dry conditions facilitated formation of crusts on whole digestate, resulting in possibility of some anaerobic release of residual  $\text{CH}_4$ . Also, in case of SID, the increased soil temperature triggered sub-soil anaerobic processes, resulting in relatively higher  $\text{N}_2\text{O}$  emissions over summer.

#### 4 Conclusions and future directions

This study demonstrates application of simultaneous *in situ* monitoring of air pollutants ( $\text{NH}_3$ ,  $\text{NO}_2$ ) and GHGs ( $\text{CH}_4$ ,  $\text{N}_2\text{O}$ ) following land application of digestate as bio fertiliser. The baseline emissions from the business as usual (BAU) have been evaluated in the first step, followed by assessment of the corresponding environmental burden minimisation potentials of three proposed mitigation measures – mixed waste digestate (MWD); soil-incorporated digestate (SID); post-methanated digestate (PMD). Our results show the proposed mitigation measures to be effective over BAU:  $\text{NH}_3$  emissions are considerably reduced from MWD and PMD (by up to 35% and 43% respectively) and significantly reduced from SID (by up to 85%). However, delayed elevation peak for  $\text{N}_2\text{O}$  in case of SID, primarily attributed to sub-surface denitrification, showed marginal increase in emissions over BAU (by up to 2%). On the other hand, PMD effectively reduced  $\text{CH}_4$  emissions (by up to 55%) with inconsiderable influence on other emissions. Nonetheless, we acknowledge these outcomes as specific to food-based digestate; the effectiveness of the proposed mitigation strategies to digestates arising from other feedstocks will vary, primarily owing to their different physico-chemical characteristics. It is also noteworthy that the results are based on digestate characteristics from our pilot-scale anaerobic reactor, which may differ for other digestate characteristics depending on the type of substrate and the type of AD process adopted.

The study highlights some paradoxical future sustainability concerns for the voluminous amounts of digestate bound to be inadvertently generated from increased processing of OFMSW using AD. It warrants timely intervention for developing adequate strategy for post-AD handling and management of digestate, ideally as a marketable product (e.g. bio fertiliser) to the urban allotment/vegetable gardeners and to the wider farming communities. In addition, it solicits consideration for more advanced methods of digestate processing and reutilisation, including - dewatering, storage, composting, curing, exploiting alternative applications in construction/regeneration activities, etc.

Further, development of a practical software tool quantifying the emissions from digestate soil applications would facilitate sustainable digestate management practice on a routine basis. In this respect, our preliminary work on meteorological consideration can serve as useful stepping stones. It is noteworthy this study reported baseline emissions from bare plots, assuming the soil amendments are meant to prepare the land for cropping. Although some literature reports on the long term emission trends, incorporating the responses from planted vegetation (see for example, Moller and Stinner 2009), these are primarily for rural setting, and a more comprehensive study on the usage of digestate as soil amendment in the urban context is recommended as a natural next step study in order to develop full appreciation of the coupled soil-vegetation effects.

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**Table 1.** List of AD parameters assessed alongside details of the analytical methods used.

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**Table 3.** Indicative field parameters showing the contrasting weather conditions during the summer and winter experiments.

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**Figure 1.** Schematic representation of the test apparatus for quantifying emissions from digestate land application, a) Spatial distribution of the three wind tunnels (plan view); b) Diagram of the wind tunnel sampling system (side view) (adopted from (Loubet et al., 1999)). [U = Upwind manifold; D = Downwind manifold].

**Figure 2.** Plot showing temporal trends in ammonia emissions to air for the four scenarios, a) Differential; b) Cumulative (expressed as percentage of total applied N). Shown alongside are the fitted regression curves.

**Figure 3.** Plot showing temporal trends in the cumulative nitrogen dioxide emissions to air for three scenarios (expressed as percentage of total applied N). Shown alongside are the fitted regression curves.

**Figure 4.** Plot showing temporal trends in the differential methane emissions to air for the four scenarios (expressed as mg C).

**Figure 5.** Plot showing temporal trends in the differential nitrous oxide emissions to air for the four scenarios (expressed as microgram N). Shown alongside is the background emission from bare soil for reference.

**Table 1.**

<b>Parameter</b>	<b>Method/Approach</b>	<b>Literature source</b>
Dry Matter (% FM)	Oven drying at 105 °C	(APHA, 2005)
Organic Matter (%)		
pH	Digital pH meter (accuracy of $\pm 0.01$ pH unit)	(APHA, 2005)
TKN in digestate (g (kg FM) <sup>-1</sup> )	Digestion of sample with H <sub>2</sub> SO <sub>4</sub> and use of Kjeldahl apparatus for distillation	(APHA, 2005)
TAN in digestate (g (kg FM) <sup>-1</sup> )	Distillation method	(APHA, 2005)
CH <sub>4</sub> , N <sub>2</sub> O in gaseous samples	Gas Chromatograph (NUCON 5700) equipped with auto sampler and a thermal conductivity detector and 6 feet long Poropak Q stainless steel column	(APHA, 2005)
NH <sub>3</sub> , NO <sub>2</sub> in gaseous samples (µg m <sup>-3</sup> )	Shimadzu spectrophotometer (UV1700)	(CPCB, 2011; Misselbrook et al., 2005b)

TKN = Total Kjeldahl nitrogen; TAN = Total available nitrogen (as ammonia)

**Table 2.**

	<b>BAU</b>	<b>MWD</b>	<b>PMD</b>
pH	8.5	8.2	8.7
Dry matter (% solids v./v.)	4.3	5.6	3.2
Organic matter (%)	18.5	16.2	15.7
Total N (g)	147.2	120.6	128.7
RAN (g)	118.8	87.8	106.5
Total S (SO <sub>3</sub> , g)	8.8	16.5	8.7
Specific gravity (kg m <sup>-3</sup> )	0.99	0.98	0.99
Fresh matter (g)	19.8	19.6	7.3
Digestate volume (L)	20	20	20

(Note: BAU - 100% OFMSW; MWD - 70% OFMSW+30% cattle dung, v./v.; PMD – matured BAU)

**Table 3.**

	<b>Avg. Air Temp. [z=5cm] (°C)</b>	<b>Avg. Soil Temp. [z=-5cm] (°C)</b>	<b>Avg. Wind Speed [z=25cm] (m s<sup>-1</sup>)</b>	<b>Soil moisture (%)</b>	<b>Precipitation(mm)</b>	<b>Qualitative comments</b>
<i>Summer experiments (Aug – Oct 2013)</i>	39	31	2	10	0.8	dry/sunny
<i>Winter experiments (Jan – Mar 2014)</i>	11	7	0.5	45	5	humid/ foggy

**Table 4.**

Feedstock type	NH <sub>3</sub>		NO <sub>2</sub>		CH <sub>4</sub>		N <sub>2</sub> O	
	<i>S</i>	<i>W</i>	<i>S</i>	<i>W</i>	<i>S</i>	<i>W</i>	<i>S</i>	<i>W</i>
BAU	5.05	4.25	0.75	0.90	1.3E-02	1.0E-02	8.5E-04	4.2E-04
MWD	2.69	1.55	0.30	0.45	1.1E-02	7.0E-03	6.3E-04	3.5E-04
SID	0.79	0.27	0.04	0.05	1.2E-02	9.0E-03	8.6E-04	5.7E-04

*S=summer (~40°C, dry/sunny); W=winter (~10°C, humid/foggy)*

Fig. 1.

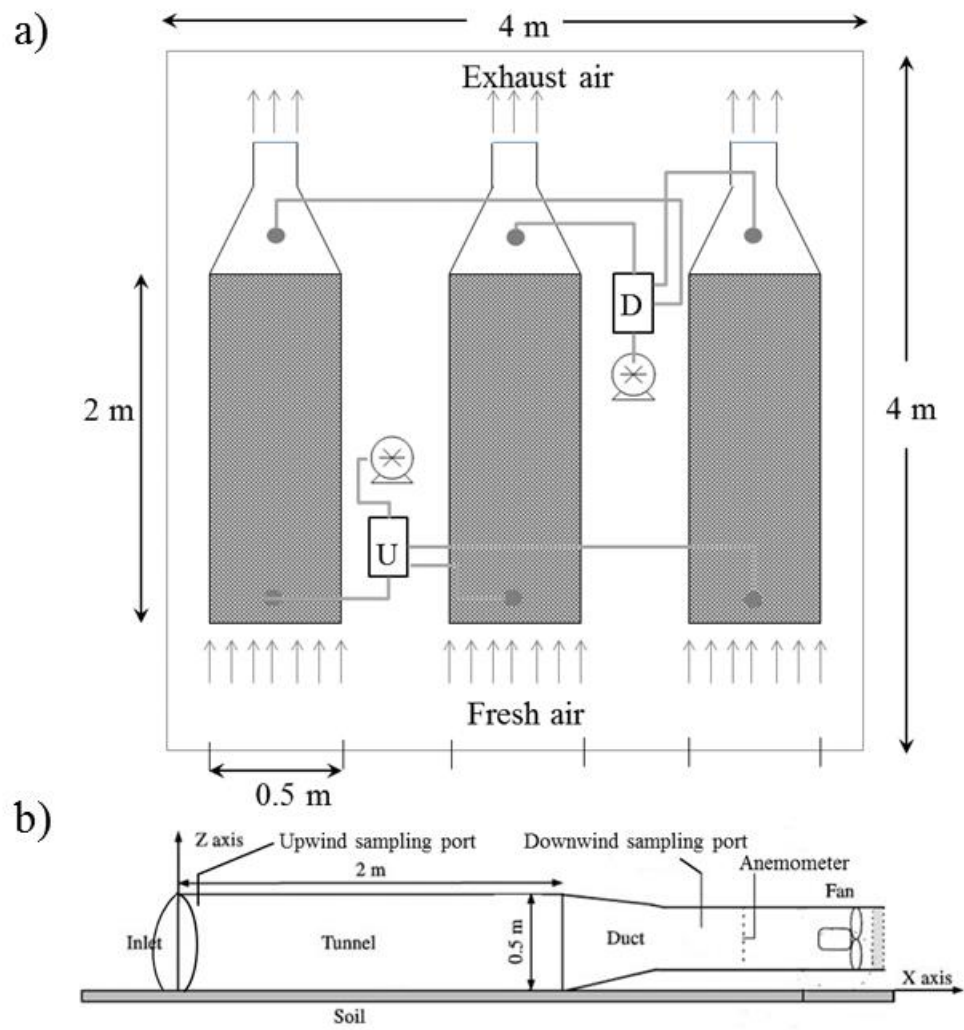
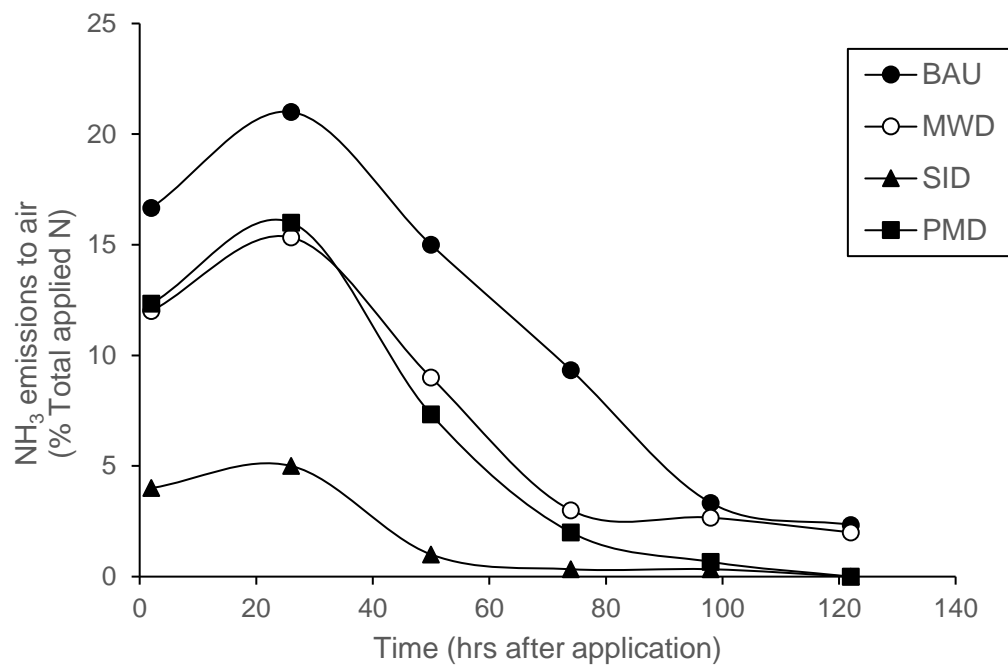




Fig. 2.

a)



b)

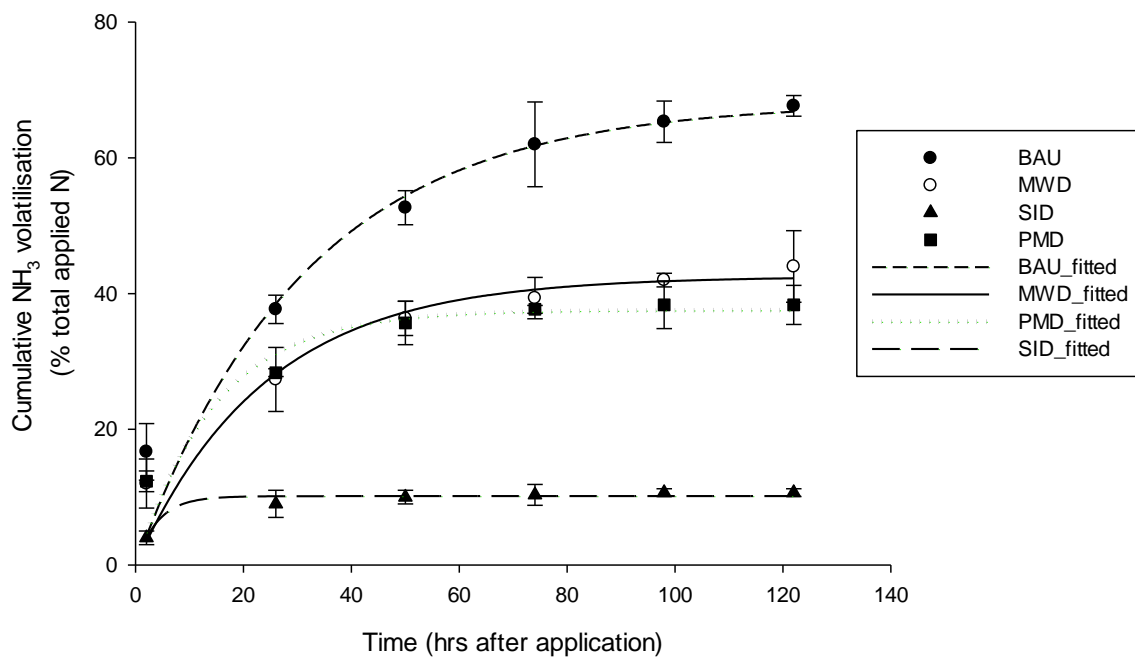


Fig. 3

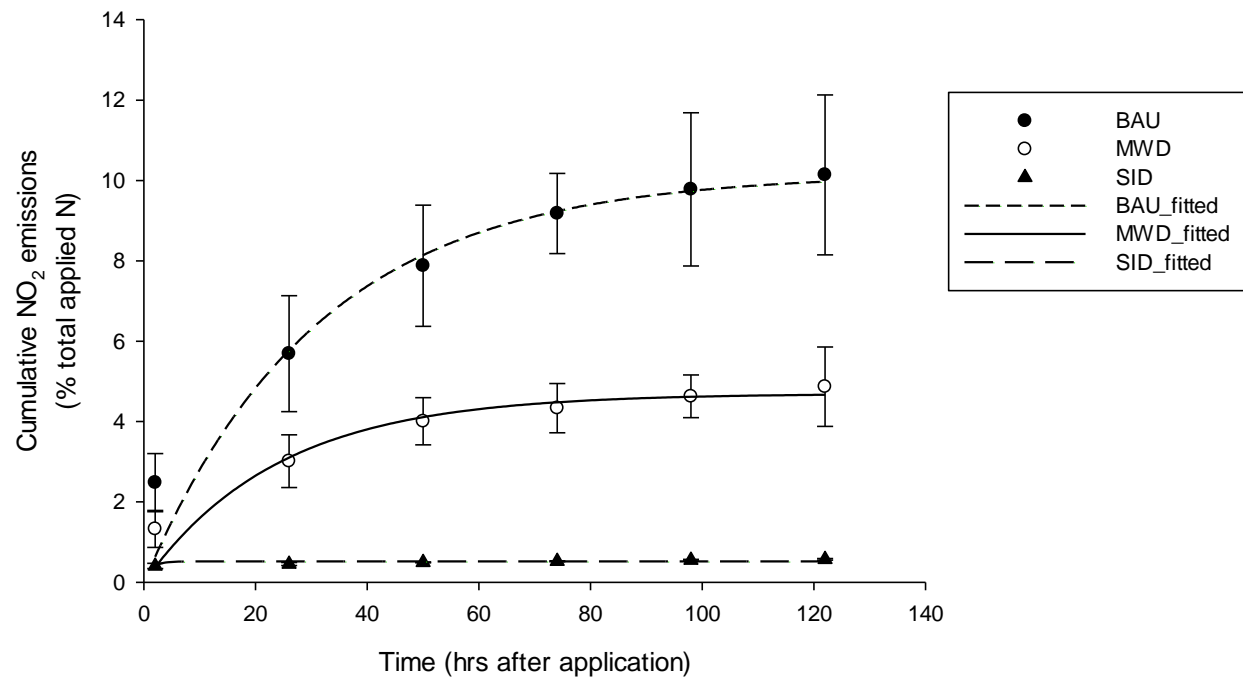


Fig. 4

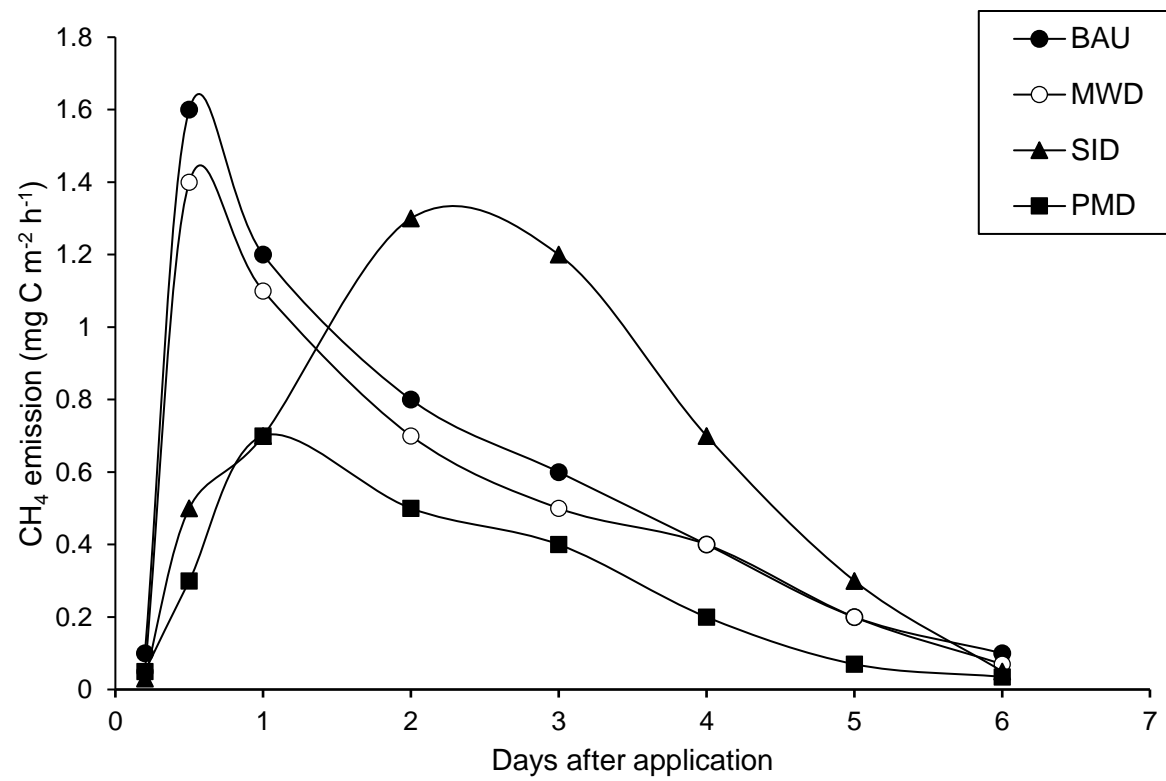


Fig. 5

